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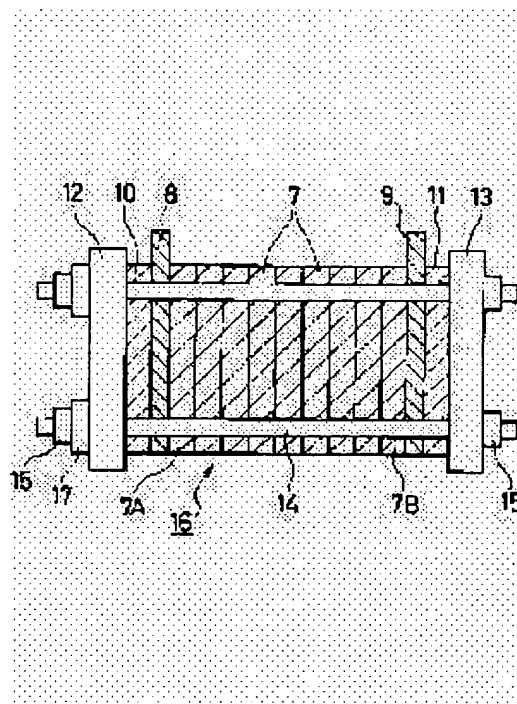
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(54) SOLID HIGH-POLYMER TYPE FUEL CELL STACK

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a solid high-polymer type fuel cell stack in which the battery performance of unit cells at the two ends of stack is prevented from dropping while temperature decrease of these unit cells is admitted.

SOLUTION: The solid high-polymer type fuel cell stack 16 is structured so that a plurality of unit cells 7 are laid one over another, wherein either of the following means is adopted, (1) the water repellency of the cathode gas diffusion layer of each unit cell located at the ends is made lower than that of the unit cells located elsewhere, (2) the gas permeability of the cathode gas diffusion layer of each unit cell located at the ends is made higher than that of the unit cells located elsewhere, (3) the specific surface area of the carbon material of the mixture layer in the cathode of each unit cell located at the ends is made greater than that of the unit cells located elsewhere, and (4) the pressure loss in the cathode side gas passage of each unit cell located at the ends is made smaller than that of the unit cells located elsewhere.



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CLAIMS

[Claim(s)]

[Claim 1] The cel unit which joined the anode to the cathode to both sides of the solid-state polyelectrolyte film, In the polymer electrolyte fuel cell stack which comes to carry out two or more laminatings of the single cel which consists of separators which have a gas passageway for being allotted to both sides of this cel unit and circulating oxidant gas or fuel gas The polymer electrolyte fuel cell stack characterized by making lower than that of a single cel which is located in other parts the water repellence of the cathode side gaseous diffusion layer of the single cel located in the both ends of this stack.

[Claim 2] The cel unit which joined the anode to the cathode to both sides of the solid-state polyelectrolyte film, In the polymer electrolyte fuel cell stack which comes to carry out two or more laminatings of the single cel which consists of separators which have a gas passageway for being allotted to both sides of this cel unit and circulating oxidant gas or fuel gas The polymer electrolyte fuel cell stack characterized by making higher than that of a single cel which is located in other parts the gas transmittance of the cathode side gaseous diffusion layer of the single cel located in the both ends of this stack.

[Claim 3] The cel unit which joined the anode to the cathode to both sides of the solid-state polyelectrolyte film, In the polymer electrolyte fuel cell stack which comes to carry out two or more laminatings of the single cel which consists of separators which have a gas passageway for being allotted to both sides of this cel unit and circulating oxidant gas or fuel gas The polymer electrolyte fuel cell stack characterized by making larger than that of a single cel which is located in other parts specific surface area of the carbon ingredient in the mixture layer in said cathode of the single cel located in the both ends of this stack.

[Claim 4] The polymer electrolyte fuel cell stack characterized by making pressure loss of the cathode side gas passageway of the single cel located in the both ends of this stack smaller than that of a single cel which is located in other parts in one polymer electrolyte fuel cell stack of claims 1-3.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] Especially this invention relates to the polymer electrolyte fuel cell stack which enabled it to prevent the cell performance degradation of a single cell located in the both ends of a stack about a polymer electrolyte fuel cell stack.

[0002]

[Description of the Prior Art] A polymer electrolyte fuel cell forms an anode and a cathode in the both sides of this solid-state polyelectrolyte film, supplies the oxidant gas (air) containing the fuel gas (hydrogen-rich gas) containing hydrogen, and oxygen, using the solid-state polyelectrolyte film as an electrolyte layer, and acquires electromotive force according to electrochemical reaction. As shown in drawing 1, an anode 2 and a cathode 3 are joined to the principal plane of the both sides of the solid-state polyelectrolyte film 1, the cell unit 4 is constituted, as for the cellular structure which is to the bases of this polymer electrolyte fuel cell, separators 5 and 6 are allotted to the both sides of this cell unit 4, respectively, and the single cell 7 is constituted.

[0003] The mixed film of the thing and fluorocarbon sulfonic acid with which said solid-state polyelectrolyte film 1 used as the cation conductivity film the polystyrene system cation exchange membrane which has a sulfonic group, and polyvinylidene fluoride, the thing which graft-ized TORIFURORO ethylene to the fluorocarbon matrix, the perfluoro carbon sulfonic-acid film (Du Pont Nafion film), etc. are known. It has the proton exchange group in the molecule, specific resistance is saturating moisture content with two or less 20-ohmcm in ordinary temperature, and these solid-states polyelectrolyte film 1 functions as a proton conductivity electrolyte.

[0004] Said anode 2 consists of anode catalyst bed 2a, gaseous diffusion layer 2b, and mixture layer 2c that intervenes among these, and a cathode 3 consists of cathode catalyst bed 3a, gaseous diffusion layer 3b, and mixture layer 3c that intervenes among these. The cell unit 4 which are an electrode / poly membrane zygote is formed by arranging and carrying out the hotpress of this anode 2 and cathode 3 to the principal plane of the both sides of said solid-state polyelectrolyte film 1.

[0005] Gas passageways 5a and 6a are formed in an inner surface side, the cooling water passage 5b and 6b is established in an outside surface side, respectively, it gets down from the separators 5 and 6 of said single cell 7, and the oxidant gas (air) to which the fuel gas (hydrogen-rich gas) containing hydrogen contains oxygen in gas-passageway 6a of the separator 6 by the side of cathode side 3 is supplied to gas-passageway 5a of the separator 5 by the side of an anode 2, respectively.

[0006] Thus, if gas is supplied, with an anode 2, electrochemical reaction to which the anode reaction which disassembles a hydrogen molecule into a hydrogen ion and an electron generates water from oxygen, a hydrogen ion, and an electron in a cathode 3 is performed, respectively, and while electromotive force arises with the electron which moves toward a cathode 3 in an external circuit from an anode 2, water will be generated at a cathode 3 side. That is, the following electrochemical reaction is performed.

Anode: $H_2 \rightarrow 2H^+ + 2e^-$ (anode reaction)

Cathode: $2\text{H}^{++} + 1/2\text{O}_2 + 2\text{e}^{-} \rightarrow \text{H}_2\text{O}$ (cathode reaction)

Whole: $\text{H}_2 + 1/2\text{O}_2 \rightarrow \text{H}_2\text{O}$ [0007] In order that the solid-state polyelectrolyte film 1 may demonstrate conductivity proper by holding to a moisture state as mentioned above, it supplies, after humidifying fuel gas and/or oxidant gas and including fixed moisture, and humidity has been made to be carried out with this humidification moisture. Moreover, although it is usually operated at 50-100 degrees C in order to make small specific resistance of the solid-state polyelectrolyte film 1 and to maintain generation efficiency highly, he supplies cooling water to the cooling water passage 5b and 6b of said separators 5 and 6, and is trying to hold said predetermined operating temperature so that temperature may not become an elevated temperature by generation of heat accompanying said electrochemical reaction.

[0008] Since the electromotive force of said single cel 7 is little, while carrying out the laminating of two or more single cels 7 in the serial direction like drawing 2 and arranging collecting electrode plates 8 and 9 on both sides, the electric insulation plates 10 and 11 are arranged on those outsides, and clamping plates 12 and 13 are further arranged on the outside, and these clamping plates 12 and 13 are bound tight with a bolt 14 and a nut 15, it fixes, and a stack 16 is constituted. In addition, 17 is a belleville spring and has given the moderate bolting force to the stack 16.

[0009]

[Problem(s) to be Solved by the Invention] In the polymer electrolyte fuel cell stack of the above-mentioned configuration, since the single cels 7A and 7B located in the both ends of a stack 16 are held where a piece side edge side is contacted to collecting electrode plates 8 and 9, as compared with the single cel 7 located in other parts by which allotted the single cel to both sides and the laminating was carried out to them, the rate which radiates heat by heat conduction from a side face becomes large. For this reason, the temperature of the single cels 7A and 7B located in the both ends of a stack 16 tends to become lower than the temperature of the single cel 7 located in other parts.

[0010] Drawing 4 was the graph which measured the cel temperature distribution in 10 cel stack which comes to carry out the ten-piece laminating of the single cel 7, and its temperature was lower than the single cel 7 to which the cel numbers 1 and 10 are the single cels located in the both ends of a stack, and both are located in other parts in this case.

[0011] Thus, if the temperature of the single cels 7A and 7B located in the both ends of a stack 16 becomes low, in the cathode 3 of these cels, the vapor pressure of electrode reaction generation water and the migration water from an anode 2 will fall, and when the water of condensation piles up in cathode side catalyst bed 3a or gaseous diffusion layer 3b, the problem of causing cell degradation will occur.

[0012] In order to solve the above-mentioned problem, there is a technique indicated by the former, for example, JP,10-308229,A. By pouring the cooling water which established cooling water passage in the electric insulation plate of a stack, passed through the inside of a stack to this cooling water passage, and carried out the temperature rise in response to the heat of a cell reaction, this conventional technique controls the heat dissipation from stack both ends, and suppresses the temperature lowering of a single cel located in both ends.

[0013] However, it is necessary to establish the exclusive path of cooling water in an electric insulation plate, and, according to this conventional technique, battery-module structure becomes complicated. Moreover, when supplying reactant gas to each cel through an internal manifold, reactant gas is usually supplied from the clamping plate side located in stack ends. When reactant gas is air, in the cel (part equivalent to 7A and 7B) in which the amount of supply is located near the clamping plate as a result of the air rate of flow within an internal manifold increasing, since [considerable] many [compared with a fuel], the amount of supply may become small compared with the cel of other parts. In such a case, even if it gives a means to control the heat dissipation from stack both ends, cell degradation may be caused by water condensation which was mentioned above.

[0014] Furthermore, although a means to attach a heat insulation member in the flank of a single cel located in the both ends of a stack, for example as other solution means, and to prevent the heat dissipation from the flank of a single cel can be considered, the configuration member of a stack

increases, it becomes cost high or difficulties, like the size of a stack becomes large arise.

[0015] Then, this invention aims at offering the polymer electrolyte fuel cell stack which enabled it to prevent the cell performance degradation of the single cel of these both ends, the configuration member of a stack admitting the temperature lowering of a single cel located in the both ends of a stack without changing.

[0016]

[Means for Solving the Problem] The cel unit with which this invention joined the anode to the cathode to both sides of the solid-state polyelectrolyte film as the 1st means in order to attain the above-mentioned object, In the polymer electrolyte fuel cell stack which comes to carry out two or more laminatings of the single cel which consists of separators which have a gas passageway for being allotted to both sides of this cel unit and circulating oxidant gas or fuel gas Let it be a summary to have made lower than that of a single cel which is located in other parts the water repellence of the cathode side gaseous diffusion layer of the single cel located in the both ends of this stack.

[0017] The cel unit which joined the anode to the cathode to both sides of the solid-state polyelectrolyte film as the 2nd means, In the polymer electrolyte fuel cell stack which comes to carry out two or more laminatings of the single cel which consists of separators which have a gas passageway for being allotted to both sides of this cel unit and circulating oxidant gas or fuel gas Let it be a summary to have made higher than that of a single cel which is located in other parts the gas transmittance of the cathode side gaseous diffusion layer of the single cel located in the both ends of this stack.

[0018] The cel unit which joined the anode to the cathode to both sides of the solid-state polyelectrolyte film as the 3rd means, In the polymer electrolyte fuel cell stack which comes to carry out two or more laminatings of the single cel which consists of separators which have a gas passageway for being allotted to both sides of this cel unit and circulating oxidant gas or fuel gas Let it be a summary to have made larger than that of a single cel which is located in other parts specific surface area of the carbon ingredient in the mixture layer in said cathode of the single cel located in the both ends of this stack.

[0019] Let it be a summary to have made pressure loss of the cathode side gas passageway of the single cel located in the both ends of this stack smaller than that of a single cel which is located in other parts in the polymer electrolyte fuel cell stack of one of the above as the 4th means.

[0020] According to said 1st means, this invention is constituted as mentioned above, and it is making low the water repellence of the cathode side gaseous diffusion layer of the single cel located in the both ends of a stack, and since it becomes easy to penetrate the produced water in a cathode, and the migration water from an anode to a gaseous diffusion layer side and a touch area with oxidant gas increases, it becomes that it is easy to be discharged with oxidant gas through a gas passageway. Into a cathode catalyst bed, water does not pile up superfluously by this, but gaseous diffusion nature becomes good.

[0021] According to said 2nd means, by making high the gas transmittance of the cathode side gaseous diffusion layer of the single cel located in the both ends of a stack, the amount of transparency of oxidant gas is increased and the produced water in a cathode and the migration water from an anode side become is easy to be discharged with oxidant gas. Into a cathode catalyst bed, water does not pile up superfluously by this, but gaseous diffusion nature becomes good.

[0022] the water absorptive power by the capillary tube force of the pore section form of a carbon ingredient particle increase by enlarge the specific surface area of the carbon ingredient in the mixture layer in the cathode of the single cel locate in the both ends of a stack, and, according to said 3rd means, for this reason, it become that produced water and migration water become easy move in the inside of a mixture layer, and stripping be carry out and it be easy be discharge in oxidant gas. Consequently, into a cathode catalyst bed, water does not pile up superfluously, but gaseous diffusion nature becomes good.

[0023] According to said 4th means, the flow rate of the oxidant gas which flows a gas passageway increases by making small pressure loss of the cathode side gas passageway of the single cel located in the both ends of a stack, and produced water and migration water become is easy to be discharged. Into a cathode catalyst bed, water does not pile up superfluously by this, but gaseous diffusion nature

becomes good.

[0024]

[Embodiment of the Invention] Next, the operation gestalt of this invention is explained. The fundamental configuration of a polymer electrolyte fuel cell stack is the same as the stack 16 shown in drawing 2, and is formed in the aforementioned way. However, the number of laminatings of the single cel 7 is set to 10, and the cel unit 4 of the single cel (it considers as the cel numbers 1 and 10) located in the both ends of a stack 16 is manufactured like the following examples.

[0025] (Example 1)

** Cut into a predetermined dimension the carbon paper (Toray Industries [, Inc.] make: TGP-H060) used as gaseous diffusion layer 3b of a cathode 3.

** The cut carbon paper is immersed in the FEP (tetrafluoroethylene-hexafluoropropylene copolymer) dispersion liquid which performed specific gravity adjustment by mixing with water. At this time, the amount of FEP was adjusted [layer / of the single cel located in the both ends of a stack 16 / gaseous diffusion] for the amount of FEP to 20% of the weight about the gaseous diffusion layer of 10 % of the weight and the other located single cels. It dries after that and heat-treats at 380 degrees C for about 1 hour.

** Form catalyst bed 3a which consists of platinum support carbon and a solid-state macromolecule on the carbon paper which ended heat treatment, and complete a cathode 3.

** Obtain the cel unit 4 by arranging and carrying out the hotpress of the anode 2 created by the existing approach, and the cathode 3 completed by ** to both sides of the solid-state polyelectrolyte film 1 (Du Pont make: Nafion112).

The water-repellent low cel unit with few [namely,] amounts of FEP of gaseous diffusion layer 3b among the cel units 4 created by the approach of ** ** - ** was used as a cel unit of the single cel located in the both ends of a stack, the cel unit with many amounts of FEP was used as a cel unit of the single cel located in other parts, the laminating was performed, and 10 cel stack was created.

[0026] This example 1 constitutes lower than the water repellence of the cathode gaseous diffusion layer of the single cel located in other parts the water repellence of cathode gaseous diffusion layer 3b of a single cel located in the both ends of a stack 16.

[0027] (Example 2)

** Cut into a predetermined dimension the carbon paper (Toray Industries [, Inc.] make: TGP-H060 thickness of about 0.2mm and TGP-H090 thickness of about 0.3mm) used as gaseous diffusion layer 3b of a cathode 3, respectively.

** These carbon paper is immersed in the FEP dispersion liquid which performed specific gravity adjustment by mixing with water. At this time, the amount of FEP was made into 20 % of the weight also in the carbon paper of which thickness. It dries after that and heat-treats at 380 degrees C for about 1 hour.

** Form catalyst bed 3a which consists of platinum support carbon and a solid-state macromolecule on the carbon paper which ended heat treatment, and complete a cathode 3.

** Obtain the cel unit 4 by arranging and carrying out the hotpress of the anode 2 created by the existing approach, and the cathode 3 completed by ** to both sides of the solid-state polyelectrolyte film 1 (Du Pont make: Nafion112).

Among the cel units 4 created by the approach of ** ** - **, the small namely, high cel unit of gas transmittance is used as a cel unit of the single cel located in the both ends of a stack 16, and the thickness of the carbon paper of cathode gaseous diffusion layer 3b uses a cel unit with large carbon paper thickness as a cel unit of the single cel located in other parts, performs a laminating, and creates 10 cel stack.

[0028] This example 2 constitutes highly the gas transmittance of cathode gaseous diffusion layer 3b of a single cel located in the both ends of a stack 16 from gas transmittance of the cathode gaseous diffusion layer of a single cel located in other parts.

[0029] (Example 3)

** Cut into a predetermined dimension the carbon paper (Toray Industries [, Inc.] make: TGP-H060)

used as gaseous diffusion layer 3b of a cathode 3.

** It is immersed in the FEP dispersion liquid which performed specific gravity adjustment by mixing with water, and dry after that and heat-treat the carbon paper (380 degrees C, 1 hour).

** Mix with 10g of carbon black (surface area 700-800m²/g) powder as a dispersant the terpineol which added the several cc surfactant for 16.7g of PTFE (polytetrafluoroethylene) dispersion liquid 60% of the weight, and create a paste.

** Mix with 10g of carbon black (surface area 200-300m²/g) powder as a dispersant the terpineol which added the several cc surfactant for 16.7g of PTFE dispersion liquid 60% of the weight again, and create a paste.

The paste obtained by ** * and ** is applied on the carbon paper obtained by **. The paste obtained by ** is used for the cel unit of the single cel located in other parts using the paste with which the cel unit of the single cel located in the both ends of a stack 16 was obtained by **.

** After drying the carbon paper which applied the paste, heat-treat at 360 degrees C for 1 hour, and complete gaseous diffusion layer 3b.

** Form catalyst bed 3a which consists of platinum support carbon (support carbon specific-surface-area 200-300m²/g) and a solid-state macromolecule on the carbon paper which ended heat treatment, and complete a cathode 3.

** Obtain the cel unit 4 by arranging and carrying out the hotpress of the anode 2 created by the existing approach, and the cathode 3 completed by ** to both sides of the solid-state polyelectrolyte film 1 (Du Pont Nafion112).

Using what has a large carbon specific surface area of the paste added to cathode gaseous diffusion layer 3b among the cel units created by the approach of ** * - ** as a cel unit of the single cel located in the both ends of a stack 16, what has a small carbon specific surface area is used as a cel unit of the single cel located in other parts, and creates 10 cel stack.

[0030] This example 3 constitutes more greatly than the single cel located in other parts the specific surface area of the carbon ingredient in mixture layer 3c in the cathode 3 of the single cel located in the both ends of a stack from a single cel located in the both ends of a stack.

[0031] (Example 4) In an example 1, in case 10 cel stack is created using the cel unit 4 used for single cels other than the both ends of a stack, 10 cel stack is created using the thing to which the channel depth of gas-passageway 6a of a separator 6 which stands face to face against the cathode 3 of the single cel located in both ends was made to increase by 10% compared with the gas passageway of a single cel located in other parts.

[0032] This example 4 constitutes small the pressure loss of gas-passageway 6a of the cathode side separator 6 of the single cel located in the both ends of a stack 16 compared with the pressure loss of the gas passageway of the cathode side separator of the single cel located in other parts.

[0033] (Example of a comparison) In order to compare with these examples 1-4, 10 cel stack was created by the same approach as an example 1 except having formed the single cel of the both ends of a stack 16 in the same specification as the single cel of other parts.

[0034] Next, the cel distribution-of-voltage trial with 10 cel stack formed in the examples 1-4 and 10 cel stack formed in the example of a comparison was performed. The test result is shown in drawing 3.

According to this test result, examples 1-4 showed the as a whole almost same distribution inclination, the value within the limits of 685-695mV was acquired, and the cel electrical potential difference of the cel numbers 2-9 was only a value slightly lower than the single cel to which the value of the range of 675-680mV is located in other parts in the single cel which is obtained and is located in the both ends of a stack 16 by 672-680mV and the cel number 10 by the cel number 1. On the other hand, although the value of within the limits as the cel numbers 2-9 in examples 1-4 with the almost same cel electrical potential difference of the cel numbers 2-9 was acquired in the example of a comparison, as compared with the cel number 1 in examples 1-4, it was far low, and by the cel number 1, it is 656mV, and was [by the cel number 10, it was 662mV, and] far low as compared with the cel number 10 in examples 1-4.

[0035] Although the cell engine performance of a single cel in which it is located in the both ends of 10

cel stack was somewhat inferior to the single cel located in other parts in the examples 1-4 from this, it became clear that it excelled farther than the single cel located in the both ends of 10 cel stack in the example of a comparison.

[0036] Since the water repellence of cathode gaseous diffusion layer 3b of a single cel located in the both ends of 10 cel stack as mentioned above was constituted from an example 1 lower than the single cel located in other parts, it becomes easy to penetrate the produced water in a cathode 3, and the migration water from an anode 2 to the gaseous diffusion layer 3b side and a touch area with oxidant gas increases, it becomes that it is easy to be discharged with oxidant gas through gas-passageway 6a. Into catalyst bed 3a of a cathode 3, water does not pile up superfluously by this, but it is thought that gaseous diffusion nature became good.

[0037] Since the gas transmittance of cathode gaseous diffusion layer 3b of a single cel located in the both ends of 10 cel stack as mentioned above was constituted from an example 2 more highly than the single cel located in other parts, Increase, the produced water in a cathode 3 and the migration water from an anode 2 side become easy to move the amount of transparency of oxidant gas with oxidant gas, and gas-passageway 6a of a separator 6 becomes is easy to be discharged. Into cathode catalyst bed 3a, water does not pile up superfluously by this, but it is thought that gaseous diffusion nature became good.

[0038] Since the specific surface area of the carbon ingredient in mixture layer 3c in the cathode 3 of the single cel located in the both ends of 10 cel stack as mentioned above was constituted from an example 3 more greatly than the single cel located in other parts, The water absorptive power by the capillary tube force of the pore section formed of a carbon ingredient particle increases. The produced water in a cathode 3 and the migration water from an anode 2 side become easy to move in the inside of mixture layer 3c, it becomes that stripping is carried out and it is easy to be discharged in oxidant gas, and water does not pile up superfluously into cathode catalyst bed 3a by this, but it is thought that gaseous diffusion nature became good.

[0039] The channel depth of gas-passageway 6a of a separator 6 which stands face to face against the cathode 3 of the single cel located in the both ends of 10 cel stack as mentioned above in the example 4 With making it deep and constituting the pressure loss of gas-passageway 6a from a gas passageway of a single cel located in other parts small The flow rate of the oxidant gas which flows gas-passageway 6a is increased, the produced water in a cathode 3 and the migration water from an anode 2 side become is easy to be discharged, and water does not pile up superfluously into cathode catalyst bed 3a by this, but it is thought that gaseous diffusion nature became good.

[0040] Thus, in this invention, it can form the stack 16 which the cell engine performance of a single cel in which it was located in both ends with the means of one of the above could be raised rather than the conventional thing, and operation of stabilized by this was attained, and was equipped with good output characteristics, admitting the temperature lowering of a single cel located in the both ends of a stack 16.

[0041]

[Effect of the Invention] As explained above, this invention is set to a polymer electrolyte fuel cell stack.

(1) the water repellence of the cathode gaseous diffusion layer of the single cel located in the both ends of a stack The gas transmittance of the cathode gaseous diffusion layer of a single cel which is constituted lower than the single cel located in other parts and which is located in the both ends of (2) stacks The specific surface area of the carbon ingredient in the mixture layer in the cathode of the single cel located in the both ends of (3) stacks highly constituted compared with the single cel located in other parts The pressure loss of the cathode side gas passageway of the single cel located in the both ends of (4) stacks constituted more greatly than the single cel located in other parts The effectiveness of making easy to discharge the produced water in the cathode in the single cel located in the both ends of a stack and the migration water from an anode side, and preventing cell performance degradation by really shifting constituting small compared with the single cel located in other parts, and adopting that means is done so.

[Translation done.]